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Preface to Ultrafast processes in chemistry and biology, a Discussion Meeting held at the Royal Society

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Preface

The Discussion Meeting was structured to provide an integrated view of the scientific questions that can now be addressed by physicists, chemists and biologists, and the experimental and computational strategies now available for studying the dynamics of atomic, molecular and biological systems on femtosecond time scales. Much of the thinking, as well as the technology, is transferable across the different physical situations and, depsite the individual groupings of the Discussion topics, it was often difficult to 'spot the join'.

The Discussion began with a survey of the generation, amplification and application of ultrashort laser pulses. The development of chirped pulse amplification has led to 'table-top' laser sources with powers in excess of 1 TW and intensities of the order of 10^{18} W cm^{-2}; these allow the generation of high harmonics into the soft X-ray region, with pulse durations of less than 1 fs. The interaction of atomic (or molecular) clusters with such high laser fields essentially creates a metal, and leads to their fragmentation into electrons and very highly charged positive ions, e.g. Xe_n^{40+} from atomic xenon clusters. The Coulomb explosion endows these ions with very high kinetic energies. The multi-electron response to very high laser fields, and the possibility of using Coulomb explosions as a means of imaging molecular structure, were intensively discussed.

The coherence of laser radiation can be exploited in many ways. Within isolated molecules, one of the most exciting possibilities is that of controlling the direction of photon-initiated chemical processes. This can be achieved through coherent phase control, either through interference through two alternative pathways (the Young's slit approach), or through 'tailoring' the phase of ultrafast laser pulses to match the wavepacket evolution in an excited molecule. In the condensed phase, one of the big surprises revealed through ultrafast laser experiments has been the realization that chemical dynamics can be followed via wavepacket motions. The dynamics are not entirely 'washed-out' by the solvent environment, though the solute species may be engaged in a vigorous 'dialogue' with their nearest neighbours. A brilliant experiment explored the consequences of charge redistribution in the hypochlorite ion (ClO^-) dissolved in an aqueous solution; electronic excitation, which transfers the negative charge from the O to the Cl atom, results in a rapid loss in the initial solvent structure and its slower subsequent re-organization to accommodate the changed field around the ion. The interaction of electronic and nuclear motion, between (and within) dissolved species and their condensed phase environments was a focal point of many contributions. Experiments can be designed to explore dynamical processes in the solute (and its environment), or, by using the solute as a probe, to explore the dynamics *in* the environment, e.g. a liquid, a glass, a photosynthetic reaction centre, or a protein. Stimulated photon echo experiments provide a means of studying and manipulating the mechanisms of spectral line broadening, which reflects the 'dialogue' between the chromophore and its environment, and determining spectral densities in the condensed phase.

In the biological field, almost all the contributions focused on the remarkable progress that has been made in the experimental and theoretical microscopic exploration of the primary process of electronic energy transfer (EET) in photosynthet-

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ic antennae and electron transfer in photosynthetic reaction centres. The ultrafast character of the EET steps is an essential component of the overall efficiency of light harvesting centres. The origin of the uni-directionality of electron transfer within the photosynthetic centre remains a puzzle.

The success of the Discussion owed much to the insight and energy of Graham Fleming, Peter Knight and Gareth Roberts (who also organized the Poster session), to the Royal Society staff for their enthusiastic support in helping to organize and publish it and, most crucially, to the authors for their cooperation in producing their contributions.

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